Thermopower of a Kondo spin-correlated quantum dot

R. Scheibner, H. Buhmann, D. Reuter†, M.N. Kiselev* and L.W. Molenkamp

*Physikalisches Institut (EP3) and *Institut für Theoretische Physik,*
Universität Würzburg, 97074 Würzburg, Germany
†*Lehrstuhl für Festkörperphysik, Ruhr-Universität Bochum, 44801 Bochum, Germany*
(Dated: February 2, 2008)

The thermopower of a Kondo-correlated gate-defined quantum dot is studied using a current heating technique. In the presence of spin correlations the thermopower shows a clear deviation from the semiclassical Mott relation between thermopower and conductivity. The strong thermopower signal indicates a significant asymmetry in the spectral density of states of the Kondo resonance with respect to the Fermi energies of the reservoirs. The observed behavior can be explained within the framework of an Anderson–impurity model.

The Kondo effect due to magnetic impurity scattering in metals is a well known and widely studied phenomenon [1]. The effect has recently received much renewed attention since it was demonstrated [2, 3] that the Kondo effect can significantly influence transport through a semiconductor quantum dot (QD). In a gate defined QD, the electronic states can be controlled externally, which allows experimenters to address many questions concerning Kondo physics [4] that were previously inaccessible. As yet unexplored are the thermoelectric properties of a QD in the presence of Kondo correlations. This is unfortunate, since these properties often yield valuable additional information concerning transport phenomena. For example, the thermopower (TP) S is related to the average energy $\langle E \rangle$ with respect to the Fermi energy E_{F} of the particles contributing to the transport by [5, 6]

$$S \equiv - \lim_{\Delta T \to 0} \left. \frac{V_{\mathrm{T}}}{\Delta T} \right|_{I=0} = - \frac{\langle E \rangle}{eT} \; , \qquad (1)$$

where V_{T} is the thermovoltage, ΔT the applied temperature difference, and e the elementary charge. S is therefore a direct measure of the weighted spectral density of states in a correlated system with respect to the Fermi energy E_F. Moreover, the TP can be used to determine the spin-entropy flux accompanying electron transport, which sets boundaries on the operating regime of spin-based quantum computers [7]. Spin entropy was recently shown to be the origin of the giant thermoelectric power of layered cobalt oxides [8], and one may anticipate similarly large effects for single quantum dots.

In this Letter we present TP measurements on a lateral QD in the presence of Kondo correlations in comparison with results obtained for the weak coupling Coulomb blockade (CB) regime. The experiments show a clear breakdown of transport electron-hole symmetry in the vicinity of Kondo spin-correlations, accompanied by de-
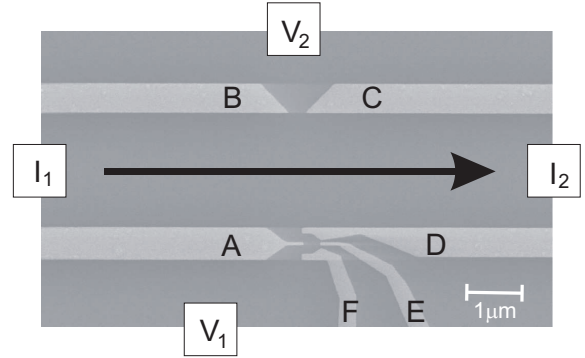

FIG. 1: SEM photograph of the quantum dot and the central part of the 20 μm long heating channel. The labels $V_{1,2}$ and $I_{1,2}$ indicate the 2DEG areas with ohmic contacts.

viations from the semiclassical Mott relation [5],

$$S_{\mathrm{Mott}} = - \frac{\pi^2}{3} \frac{k^2 T}{e} \frac{\partial \ln G(E)}{\partial E}, \qquad (2)$$

where k_B is Boltzmann's constant, and $G(E)$ is the energy-dependent conductance of the QD.

Figure 1 shows an SEM-photograph of the sample structure. This structure is fabricated by electron-beam lithography on a (Al,Ga)As/GaAs heterostructure containing a two dimensional electron gas (2DEG) with carrier density $n_s = 2.3 \times 10^{15}$ m^{-2} and mobility $\mu = 100$ m^2/(Vs). Gates A, D, E, and F form the QD and gates A, B, C, and D are the boundaries of the electron-heating channel, which is 20 μm long and 2 μm wide. The QD has a lithographical diameter of approximately 250 nm and has a design [9] that allows great flexibility in the number of electron on the dot. For the present experiment, the barriers are adjusted such that the number of electrons can be varied conveniently (i.e., by changing only the voltage V_{E} applied to plunger gate E) between 20 and 40, as can be deduced from the magnetic-field evolution of the CB peaks. The sample is mounted in
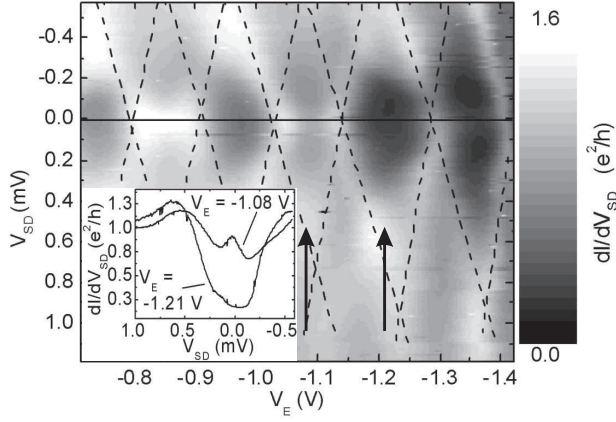
FIG. 2: Grey-scale plot of the differential conductance as a function of the QD potential ($\propto V_E$) and the externally applied bias voltage across the QD (V_{SD}). Alternating regimes of low and high conductance are observed between each successive conductance peaks within the CB diamonds (dashed lines). Inset: Bias-voltage (V_{SD}) depending traces of the differential conductivity of the dot for $V_E = -1.08$ V and $V_E = -1.21$ V [indicated by arrows].

a top loading dilution cryostat with a base temperature T_{bath} below 50 mK. For conductance measurements gates B and C are grounded. Figure 2 shows a grey-scale plot of the differential conductance of the QD as a function of its potential ($\propto V_E$) and the bias voltage across the dot (V_{SD}). Along the zero bias line [$V_{SD} = 0$, solid line in Fig. 2], alternating regimes of low and high conductance in between two successive conductance peaks are observed in the gate-voltage range $-0.7 < V_E < -1.2$ V. A V_{SD}-dependent trace [inset of Fig. 2] for $V_E = -1.08$ V (high conductance between two main CB peaks) shows a clear zero bias resonance, in contrast to a V_{SD} trace taken in the adjacent conductance valley ($V_E = -1.21$ V).

Such a zero bias resonance is characteristic for a Kondo correlated system [1]. In the following, we assign the regions around $V_E \approx -0.85$ V and -1.08 V as voltage ranges where spin correlations modify the transport characteristics. For V_E more negative than -1.2 V the coupling between the QD electrons and the surrounding 2DEG decreases and the Kondo correlations are suppressed. From temperature dependent measurements and the analysis of the CB diamonds [10] we deduce charging energies varying from $E_C = 0.7$ meV to $E_C = 1.5$ meV and corresponding intrinsic level widths of $\Gamma \sim 0.35$ meV to $\Gamma \sim 0.15$ meV in the regions of strong and weak coupling of the dot to the reservoirs, respectively.

Experimentally, the main problem in performing TP measurements on nanostructures is how to obtain a sizable temperature difference across a very small device. Here, we use a current heating technique [11, 12], that previously has been applied to measure the TP of metallic QDs [13]. A small ac-heating current is passed through the channel defined by gates A, B, C, and D. Energy dis-

sipation in the channel, results in a local heating of the electron gas. Due to the small electron-phonon coupling in (Al,Ga)As 2DEGs at low temperatures, hot electrons can only dissipate their excess energy to the lattice in the wide 2DEG area behind the channel exit, while rapid electron-electron scattering ensures thermalization of the electrons in the channel to a temperature T_{ch} which is higher than the lattice temperature T_L. Hence, the QD is embedded between the hot electron reservoir in the channel (with electron temperature $T_{el} = T_{ch}$) and the cold surrounding 2DEG ($T_{el} = T_L$). The constant temperature difference of $\Delta T = T_{ch} - T_L$ across the QD can be adjusted via the current through the channel. For the experiments presented in Figs. 3 and 4(a), we use a heating current of $I_H = 4.2$ nA. This results in $\Delta T \approx 10$ mK, where ΔT is calibrated by making use of the quantized TP of the quantum point contacts formed by gates B and C (QPC$_{BC}$) and A and B [12]. (For this calibration the QD, gates E and F are set to ground.)

The thermovoltage across the QD is measured as the voltage difference $V_T = V_1 - V_2$ (see Fig. 1). V_T contains the TP of the QD (S_{QD}) as well as that of QPC$_{BC}$ (S_{QPC}). For convenience, the TP of QPC$_{BC}$ is adjusted to zero by setting its conductance at the center of a conductance plateau. Figure 3 presents our results on V_T as a function of gate voltage V_E (lower panel) together with the corresponding conductance curve (upper panel). As discussed above, for gate voltages less negative than -1.1 V a strong coupling of the QD to the leads results in Kondo correlations at $V_E \approx -0.72$ V and -0.95 V, indicated by the horizontal bars underneath the top (conductance) panel. [Note that these values have shifted with respect to those in Fig. 2 because of the additional negative gate voltages on gates B and C] In the bottom (thermovoltage) panel, a striking difference between the behavior of V_T in the spin correlated regime as compared with the weak coupling regime is directly conspicuous.

In the weak coupling regime the line shape of the thermovoltage has negative and positive contributions in the vicinity of a conductance peak. As shown in Ref. [14], this behavior results from contributions of two different transport mechanisms: (1) the linear increase in thermovoltage at the center of a conductance peak is characteristic of sequential tunneling, while (2) its rapid fall-off between two conductance peaks is a consequence of cotunneling processes. In this regime, the thermovoltage line-shape follows qualitatively the negative parametric derivative of the conductance data, as described by Mott's relation, Eq. (2) [15, 16]. The grey line in the bottom panel of Fig. 3 shows the behavior of the thermopower as expected from Mott's relation.

The striking experimental result for the strong coupling regime is now that in the presence of spin correlations at $V_E = -0.72$ V and -0.95 V, V_T exhibits *only positive* values, while when these correlations are absent ($V_E = -0.62$ V and -0.83 V) the usual nega-
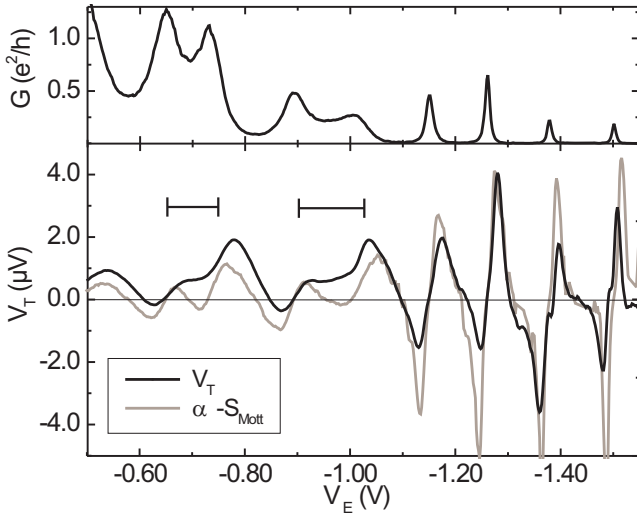
FIG. 3: Comparison of the thermovoltage measurement (V_T), the conductance (G), and the calculated thermopower S_{Mott} as expected from the Mott-relation [Eq. (2), where $E \propto (-eV_E)$] as a function of applied gate voltage V_E. The horizontal bars indicate the regions of spin correlations within the gate voltage range of strong coupling of the quantum dot to the leads ($V_E > -1.1$V).

FIG. 4: (a) Thermovoltage signal for various lattice temperatures at constant ΔT. The curves are normalized to the value at $V_E = -1.8$ V. At high temperatures the spin contribution to the thermopower between two CB peaks decreases and the oscillating CB substructure of the thermopower reappears (indicated by the arrow). (b,c) Temperature dependent (differential) conductance of the zero bias resonance as a function of bias voltage (V_{SD}) and QD potential (V_E).

tive V_T shows up in between the Coulomb resonances. A comparison with the semiclassically expected S_{Mott} [c.f. Eq. (2)] in Fig. 3 shows at the additional contributions at $V_E = -0.72$ V and -0.95 V an amount of ca. 0.5 μV, which thus cannot originate from single particle effects. Clearly, spin correlations are a prime candidate for explaining the occurrence of these extra contributions.

Further information can be obtained from the dependencies of these effects on lattice temperature T_L, which are shown in Fig. 4. [Note that these experiments were done on the same sample but for a different cooling cycle, where the regime of spin correlations was observed for a different adjustment of the voltages applied to the QD gates.] In Fig. 4(a) we plot the temperature dependence of the TP anomaly. We observe that the additional contributions to the TP are suppressed at higher temperatures. As a consequence, the expected valley reappears between the main CB resonances. For comparison we show in Figs. 4(b) and (c) the temperature evolution of the bias(V_{SD})-dependent differential conductance and the zero bias conductance of the QD. We observe that the zero bias resonance in these curves disappears on a similar temperature scale as the extra features in the TP, establishing a very strong indication that these features are related to spin correlations.

The anomalous behavior of the TP in the spin-correlation regime points to an asymmetry in the electron-hole transport, which can be understood as follows: In a TP measurement the electron heating channel has a higher temperature and therefore more empty
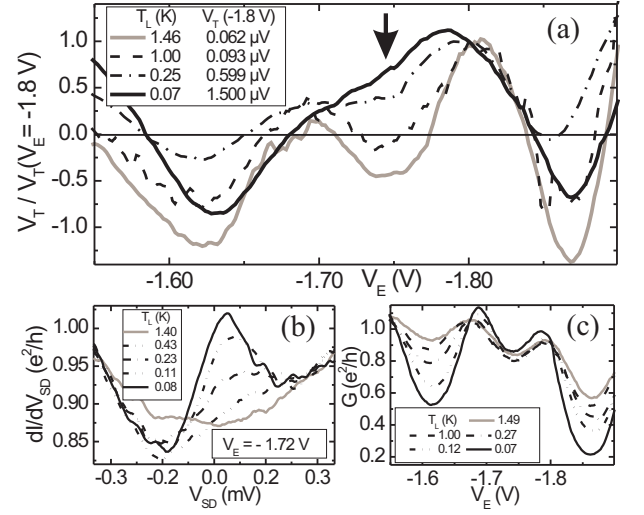
(occupied) states below (above) E_F than the colder surrounding 2DEG. When a QD level has an energy ε_{QD} slightly higher than the Fermi level E_F of the reservoirs ($E_F < \varepsilon_{QD} < E_F + k_B T$), the dominating transport process is electron-like: an electron first tunnels onto the QD and then out again into the other lead. The average energy $\langle E \rangle$ of the charges moving from the hot reservoir to the cold reservoir is positive with respect to E_F [14]. Correspondingly, if $E_F > \varepsilon_{QD} > E_F - k_B T$, $\langle E \rangle$ is negative referred to E_F and the charge transport can be described as a hole-like process. Hence, one can directly deduce the relative position of ε_{QD} to E_F from the sign of the TP signal, as well as infer whether the contributing transport process is electron- or hole-like.

In order to explain our observation of a positive thermovoltage signal, the above reasoning directly implies that the spectral density of the correlated state on the QD must have its weighted maximum above E_F in the leads. From the experimental characterization of our QD, we find that $\varepsilon_{QD}/\Gamma > -3$ in the gate voltage region where we observe spin correlations [c.f. Fig. 3 ($V_E > -1.2$ V)]. By describing the spin correlated QD in terms of an asymmetric Anderson model (i.e. $\varepsilon_{QD} \neq -E_C/2$ [20]) [1, 17, 18, 19], this implies that we are in the mixed-valence regime. In this limit, the spectral density of the hybridized state has its maximum above E_F [1, 17]. This is because strong coupling to the reservoirs leads to significant charge valence fluctuations, i.e. a delocalization of the QD charge, which results in an overall asymmetry between electron and hole like transport. One thus

expects enhanced positive contributions to the thermovoltage [17]. This is in contrast to numerical calculations for an ideal Kondo-QD system, where one expects both positive and negative contributions to the TP [19].

We note here that Kondo QDs reported on experimentally very often operate in the mixed valence regime [2, 3]. Because of the insensitivity of zero bias conductance measurements to the exact location of the spectral density of the hybridized state, this notion may have escaped general attention. The TP is in contrast very sensitive to this location [18], which is why we observe positive contributions to the TP. Further deviations of the experiment from the single level Anderson model occur due to the intrinsic level broadening of the QD states, which even leads to an overlap of the states across the CB gap, resulting in a non-zero conductance in the CB valleys without spin correlations at $V_E > -1.1\text{V}$ (c.f. Fig. 3).

Finally, we may verify the nature of the entropy flux driving the thermovoltage. In principle, one expects contributions from both spin and orbital degrees of freedom to the entropy flux [21]. However, for strongly correlated systems the spin degrees of freedom should dominate the TP [6]. In the spin correlated regime, the spin degeneracy on the QD opens a channel for spin entropy transport between the reservoirs. According to Heikes' formula [6], $S = -\sigma/e$, where σ is the entropy change during the transport of a single particle through the QD, we estimate a spin entropy contribution to the TP of $S_s = (k_B/e)\ln 2 \approx -60\ \mu\text{V/K}$. From the measurements (Fig. 3) we find an additional thermovoltage at the resonances between the CB peaks at $V_E = -0.72$ V and -0.95 V of $V_T \sim 0.5\ \mu\text{V}$ which yields a TP of $S \approx -50\ \mu\text{V/K}$ using Eq. (1) and $\Delta T \approx 10$ mK. This value agrees very well with the expected value for S_s and shows that the effect we measure is strongly spin-entropy driven. This observation provides independent and direct evidence of the correlated character of the transport through the Kondo QD. Previously, such direct information has only been obtained in bulk materials, such as 1-dimensional organic salts [21] and, recently, in cobalt oxides [8].

In summary, we have measured the TP of a small QD. The TP exhibits an additional contribution due to spin entropy when the dot is tuned into the spin correlated (Kondo) regime. The comparison of the TP in the pure CB regime with the TP in the spin-correlated regime reflects a lifting of the symmetry in the electron-hole transport. The semiclassical Mott relation between the TP and the conductance fails to describe this asymmetry, which is due to the many-particle nature of the correlation induced resonance. The measurements agree with theoretical considerations [17, 18] addressing the evolution of the TP of a Kondo correlated system as a function of QD energy. The presented results confirm that the spectral density of states for the Kondo-resonance of a dot can be explained in the framework of an Anderson impurity model in the presence of charge fluctuations. Our observation demonstrates that it is possible to create correlated thermoelectric transport in man-made nanostructures, where the experimenter has a close control of the exact transport conditions. Future detailed studies will address the quenching of the spin correlation in in-plane magnetic fields, in analogy to Ref. [8], as well as the magnitude and scaling behavior of the spin correlation contribution to the TP in comparison with results of renormalization group calculations.

[1] A.C. Hewson, *The Kondo Problem to Heavy Fermions, Cambridge Studies in Magnetism*, (Cambridge University Press, Cambridge, England, 1993).

[2] D. Glodhaber-Gordon, *et al.*, Nature **391**, 156 (1998).

[3] S. M. Cronenwett, T.H. Oosterkamp, and L.P. Kouwenhoven, Science **281**, 540 (1998).

[4] W.G. van der Wiel, *et al.*, Science **289**, 2015 (2000).

[5] J.M. Ziman, *Electrons and Phonons*, Oxford University Press (1960).

[6] P.M. Chaikin and G. Beni, Phys. Rev. B **13**, 647 (1976).

[7] D. Loss and D.P. DiVincenzo, Phys. Rev. A **57**, 120 (1998).

[8] Y. Wang, N. S. Rogado, R.J. Cava and N.P. Ong, Nature **423**, 425 (2003).

[9] M. Ciorga, *et al.*, Phys. Rev. B **61**, R16315 (2000).

[10] L.P. Kouwenhoven, *et al.*, in *Mesoscopic Electron Transport*, edited by H. Grabert, J.M. Martinis, and G. Schön, NATO ASI, Ser. E (Kluwer Dodrecht, Netherlands, 1997), and references therein.

[11] L.W. Molenkamp, *et al.*, Phys. Rev. Lett. **65**, 1052 (1990).

[12] H. van Houten, L.W. Molenkamp, C.W.J. Beenakker, and C.T. Foxon, Semicond. Sci. Technol. **7**, B215 (1992).

[13] S. Möller, H. Buhmann, S.F. Godijn, and L.W. Molenkamp, Phys. Rev. Lett. **81**, 5197 (1998); S.F. Godijn, *et al.*, Phys. Rev. Lett. **82**, 2927 (1999).

[14] M. Turek and K.A. Matveev, Phys. Rev. B **65**, 115332 (2001); A.V. Andreev and K.A. Matveev, Phys. Rev. Lett.**86**, 280 (2001).

[15] Note that Mott's relation is not quantitatively valid in the sequential tunneling regime. See also ref. [16].

[16] R. Scheibner, *et al.*, in preparation.

[17] T.A. Costi, A.C. Hewson, and V. Zlatić, J. Phys.: Condens. Matter **6**, 2519 (1994).

[18] D. Boese and R. Fazio, Europhys. Lett. **56**, 576 (2001).

[19] B. Dong and X.L. Lei, J.Phys.: Condens. Matter **14**, 11747 (2002).

[20] Note that using the notation of the Anderson model as described in Refs. [1] and [17], ε_{QD} varies between 0 and $-E_C/2$ during the CB oscillations.

[21] P.M. Chaikin, J.F. Kwak, and A.J. Epstein, Phys. Rev. Lett. **42**, 1178 (1979).